



Much work remains in order to define the chemical, as opposed to the elemental, composition, especially for organic compounds.

At nonurban eastern U.S. sites, a large fraction of the fine aerosols are composed of sulfate and related species (ammonium ions, hydrogen ions, and associated water) and organic compounds. In the northeastern and southeastern U.S., organic carbon appear to equal sulfate in the fourth quarter of the year. In the southwestern U.S., wind blown dust is a major component of fine mass while sulfate is less important (Schichtel and Husar, 1991).

Annually averaged fine particle sulfate, as ammonium sulfate; organic carbon; elemental carbon; and nitrate, as ammonium nitrate, concentrations from the IMPROVE network across the U.S. are shown in Figures 6-12 and 6-13 (Sisler et al., 1993; Malm et al., 1994b). The station density is limited, especially in the eastern U.S. The contour lines in the annual average maps are to be used as guides to the eye, rather than precise values. Concentrations of sulfate in the eastern U.S. (Figure 6-12a) exceeds those over the mountainous western states by factor of five or more. Elevated sulfate in excess of  $1 \mu\text{g}/\text{m}^3$  is also reported over the Pacific coast states. Sulfates typically contribute over 50% of the fine particulate mass in the eastern U.S., while sulfates contribute 30% or below in the West.

Fine particle nitrates (Figure 6-12b) are highest in California, exceeding  $4 \mu\text{g}/\text{m}^3$  at most sites. Their share of the fine mass at several California sites exceeds 20%. Organic carbon concentrations (Figure 6-13a) are high over California and northwestern sites, as well as at the eastern U.S. sites. Organic carbon contributes over 50% of the fine particle mass in the Northwest, and about 30% throughout the eastern U.S. There is a high degree of uncertainty associated with the measurement of particulate nitrate and organic carbon because of artifacts arising from the adsorption of vapors or the loss of semivolatile materials. The elemental carbon concentrations (Figure 6-13b) are significant over the Northwest and southern California, as well as at the Washington, DC, site. Over most of the country elemental carbon is 5% or less of the fine particle mass.

The chemical composition of  $\text{PM}_{10}$  and  $\text{PM}_{2.5}$  aerosols in the IMPROVE network (Eldred et al., 1994b) revealed that the average coarse mass does not differ significantly between the East and West; however, the fine mass is higher in the East. Also about 80% of

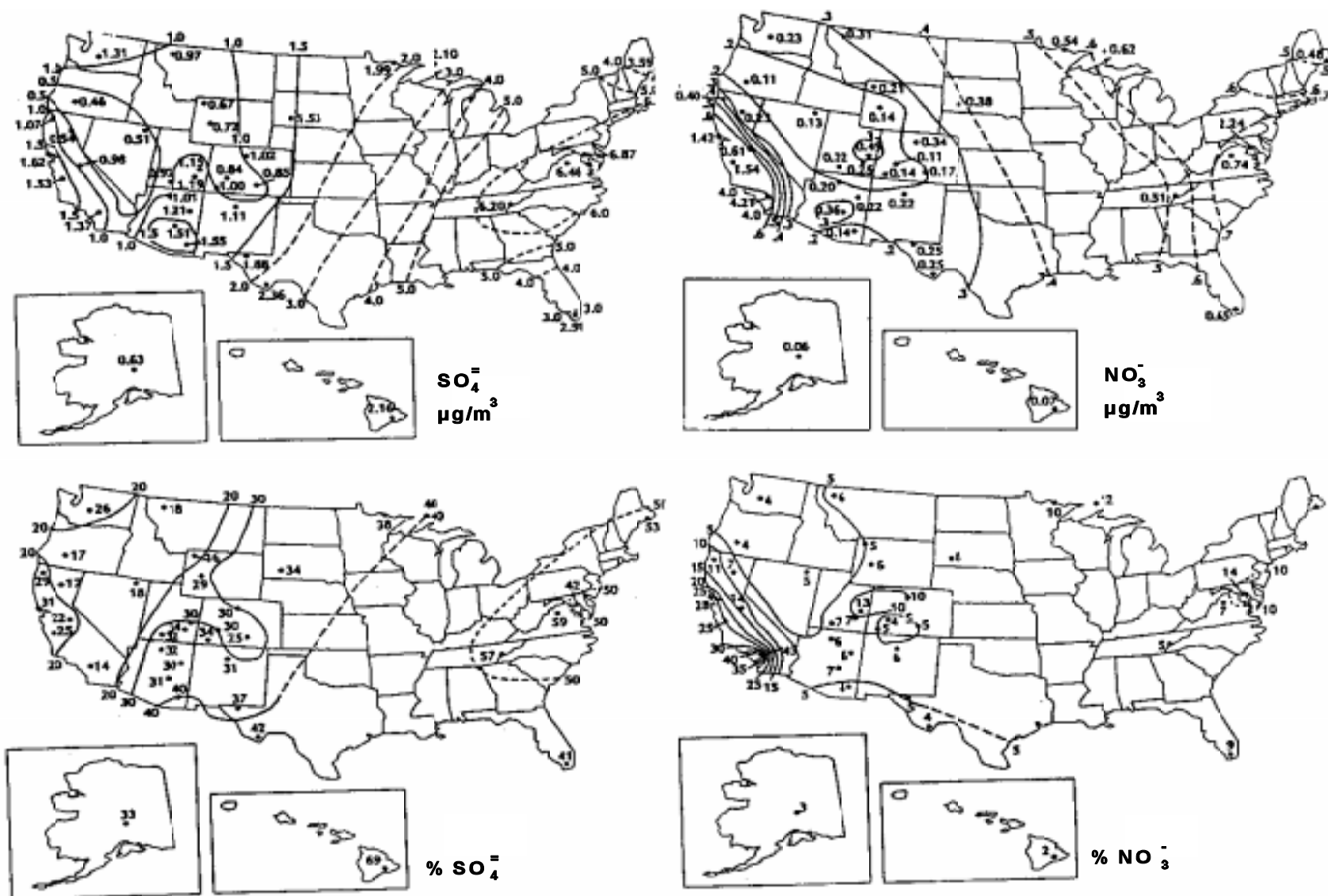


Figure 6-12. Yearly average absolute and relative concentrations for sulfate and nitrate.

Source: Sisler et al. (1993) and Malm et al. (1994b).

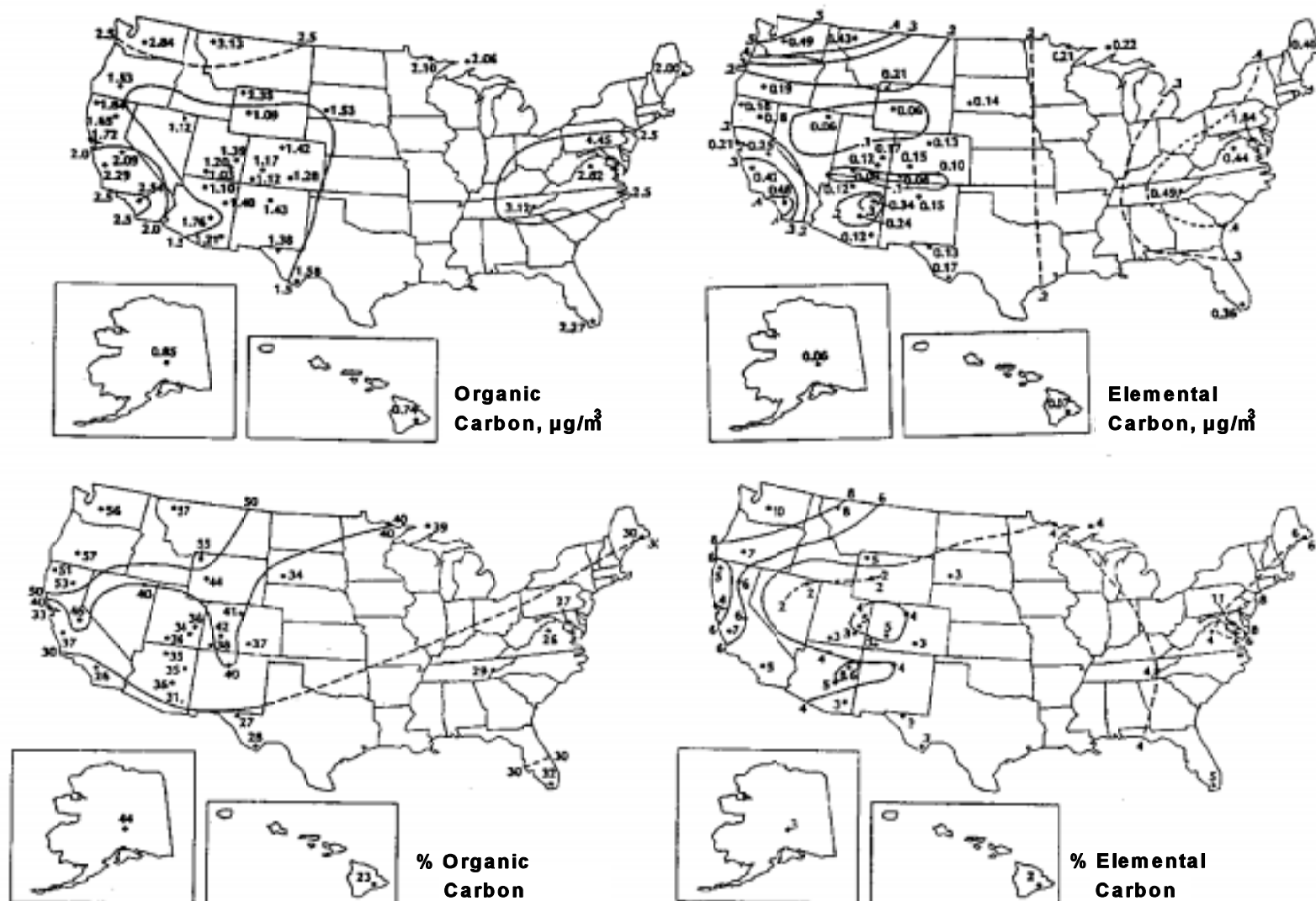


Figure 6-13. Yearly average absolute and relative concentrations for organic carbon and elemental carbon.

Source: Sisler et al. (1993) and Malm et al. (1994b).

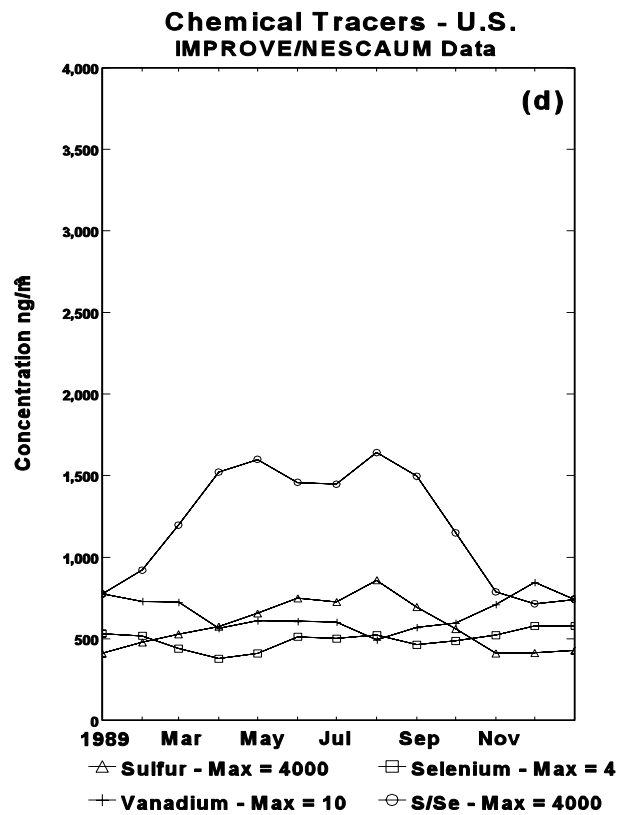
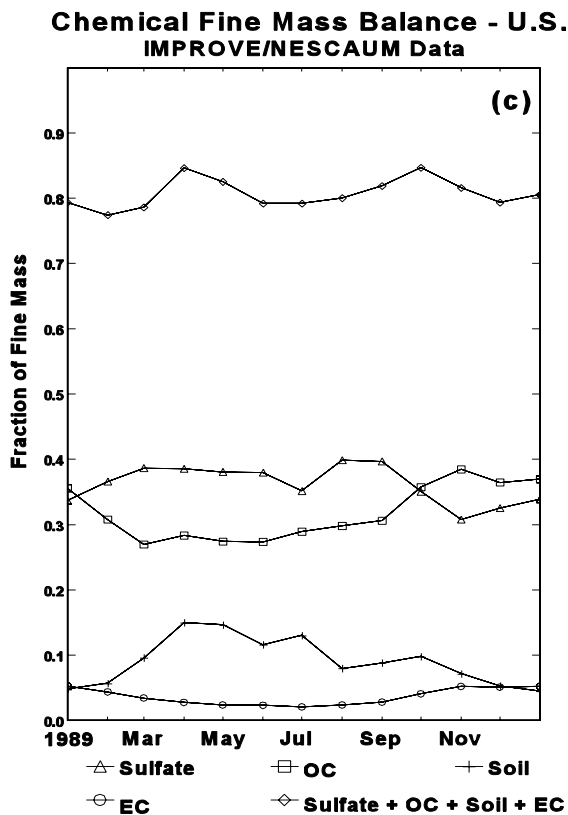
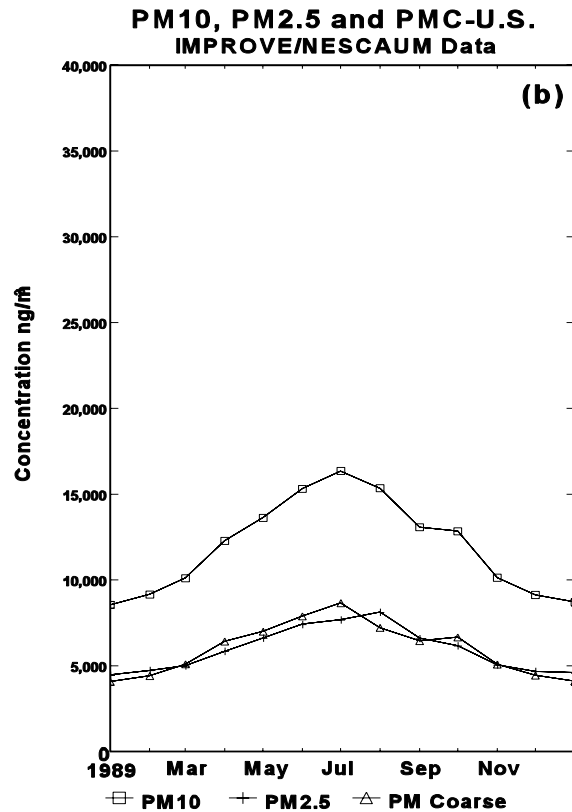
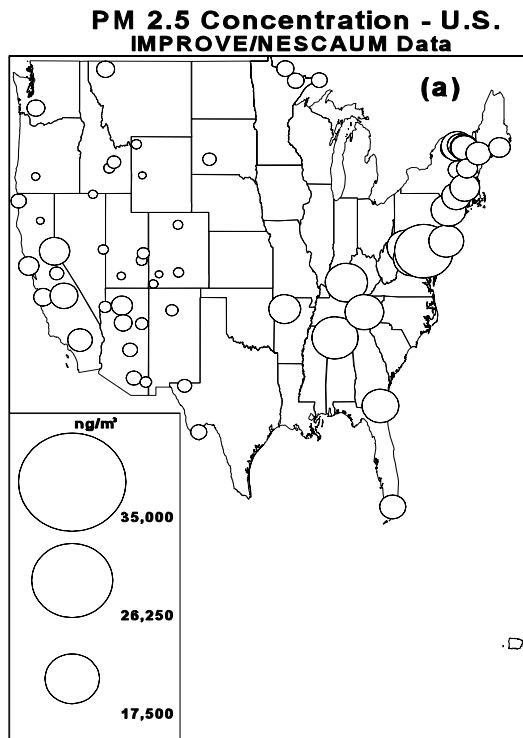
soil elements and 20% of sulfur were found in the coarse fraction. Most trace elements were found in the fine fraction, both in the East and in the West. The spatial and seasonal patterns in particle concentrations and their relationships to optical extinction in the United States from the IMPROVE network were summarized by Malm et al. (1994b).

In studying the regional patterns of nonurban trace metals in the IMPROVE network, Eldred et al. (1994a) found a good correlation between selenium and sulfur at all sites in the East. The correlation in the West is lower. Comparison of the S/Se ratios for summer and winter shows that there is approximately twice the sulfur relative to selenium in summer compared to winter. Se is a tracer for S emitted from coal-fired fossil fuel power plants; this shift in S/Se from summer to winter is consistent with a substantial secondary photochemical contribution to  $\text{SO}_4^{=}$  during the summer. Zinc is highest at the sites in the central East. It does not correlate well with sulfur. Lead and bromine are relatively uniform, with slightly higher mean concentrations in the East. There is poor correlation between lead and bromine. Copper and arsenic are highest in the Arizona copper smelter region. Copper is also higher in the central East.

Trends (1982 to 1992) of nonurban fine particle sulfur, zinc, lead, and soil elements were reported by Eldred et al. (1994a) using the IMPROVE network data. They observe that in the southwest, sulfur trends in spring, summer, and fall decreased, while most of the winter trends increased. The trends in the Northwest increase slightly. The two eastern sites (Shenandoah and Great Smoky Mountains) have increased almost 4% per year in summer, increased 1 to 3% in spring and fall, and decreased 2% in winter. The annual increase was between 2 and 3%. Generally, there were no significant trends in zinc and the soil elements. Lead at all sites decreased sharply through 1986, corresponding to the shift to unleaded gasoline. The ten year trends reported by Eldred et al. (1994b) have not been compared and reconciled with other compatible data.

#### **6.3.1.6 Seasonality of the Nonurban Chemistry**

This section discusses the seasonality of size segregated chemical composition at non-urban monitoring sites (IMPROVE/NESCAUM) over the entire U.S. (Figure 6-14).



**Figure 6-14. Seasonal pattern of nonurban aerosol concentrations for the entire United States: (a) monitoring locations; (b) PM<sub>10</sub>, PM<sub>2.5</sub>, and PMCoarse (PMC); (c) sulfate, soil, organic carbon (OC), and elemental carbon (EC) fractions; and (d) tracers.**

The nationally aggregated average  $PM_{10}$ ,  $PM_{2.5}$  and  $PM_{Coarse}$  is shown in Figure 6-14b (See Section 6.3.1.2 for a definition of  $PM_{Coarse}$ ). The nonurban  $PM_{10}$  concentration ranges from  $8 \mu\text{g}/\text{m}^3$  in the winter, December through February, to about  $15 \mu\text{g}/\text{m}^3$  in June to August. On the national scale the  $PM_{10}$  seasonality is clearly sinusoidal with a summer peak. Fine particles over the nonurban conterminous United States account for about 50 to 60% of the  $PM_{10}$  mass concentration throughout the year. The coarse mass accounts for 40 to 50% throughout the year. Hence, the fine-coarse aerosol ratio does not change dramatically for the average nonurban aerosol.

The relative chemical composition of the aggregated nonurban aerosol pattern is shown in Figure 6-14c, including sulfates, organic carbon, soil, and elemental carbon as a fraction of the fine particle mass concentration. The Figure also shows the sum of these four aerosol species to indicate the fraction of the fine aerosol mass that is not accounted for. Most notable among the missing species is the contribution of nitrates, ammonium ion, and hydrogen ion.

There is mild seasonality in the nationally aggregated sulfate and organic carbon fractions. Throughout the year, sulfate aerosol, including the ammonium cation, accounts for 30 to 40% of the fine mass. Organic carbon also contribute 30 to 40% of the nationally averaged fine particle mass. Thus, sulfates and organic carbon are the two dominant species, contributing about 70% of the fine aerosol mass.

The contribution of soil dust to the fine mass ranges between 4% in the winter months to 12% during April through July. Elemental carbon is about 2% during the summer and 5% during the winter.

The sum of the four measured fine mass components, sulfates, soil, organic carbon, and elemental carbon add up to about 80% of the measured fine mass throughout the year. The remaining, unaccounted fine mass may be contributed by nitrates, trace metals (e.g., Pb, Br, sea salt [NaCl], etc.).

The seasonal pattern of concentration of primary emission tracers, selenium, Se and vanadium, V is shown Figure 6-14d. Se is a known tracer for coal combustion, while V is a trace constituent of fuel oil (Altshuller, 1980; Kleinman et al., 1980; Cass and McRae, 1983; Tuncel et al. 1985). The Figure also shows the monthly average concentration of fine particle sulfur as well as the S/Se ratio.

The national average Se concentration is rather uniform over the seasons, ranging between 400 to 600 pg/m<sup>3</sup>. Since Se is a primary pollutant, the seasonal invariance means that the combined effect of emissions and dilution is seasonally invariant over the year.

The concentration of V is between 500 to 700 pg/m<sup>3</sup>, with the higher concentrations occurring in the winter season. Evidently, the contributions from V-bearing fuel oil are more pronounced during the cold season. The monthly average sulfur in aerosols exhibits the highest concentrations 1.5 μg/m<sup>3</sup>, during June, July, and August, and the lowest values 0.9 μg/m<sup>3</sup>, during November, December, and January.

The S/Se mass ratio is about 700 during November to January and climbs to about 1,500 during April through September. The higher S/Se ratio during the warm season is an indication of secondary sulfate production from SO<sub>2</sub> in the plumes of coal fired power plants (Chapter 3).

### **Eastern United States**

The seasonal pattern of the eastern U.S. aerosol chemistry is shown in Figure 6-15. The concentration of PM<sub>10</sub>, PM<sub>2.5</sub>, PMCoarse (Figure 6-15b) indicates a similar seasonality, highest concentrations in the summer, and lowest in the winter. The PM<sub>10</sub> levels range between 12 to 24 μg/m<sup>3</sup>, the PM<sub>2.5</sub> ranges between 8 to 12 μg/m<sup>3</sup>, while PMCoarse ranges between 4 to 7 μg/m<sup>3</sup> over the year. The size segregated aerosol data for the nonurban East show that the fine mass concentration (8 to 12 μg/m<sup>3</sup>) is higher than the national average (4 to 8 μg/m<sup>3</sup>), while the coarse mass concentration is comparable to the national average. Eastern U.S. nonurban fine particles contribute 60 to 70% of the fine mass throughout the year.

The apportionment of the fine particle mass into its chemical components (Figure 6-15c) favors sulfates which amount to 40 to 50% of the fine mass throughout the year, compared to about 30% of organic carbon. The contribution of soil dust is about 5% throughout the year, while soot is more important in the winter (6%) than in the summer (3%). The above three aerosol chemical components account for 85 to 90% of the measured fine particle mass, leaving only relatively small contribution to nitrates, hydrogen ions, trace metals, and sea salt.

The coal tracer selenium (Figure 6-15d) exhibits a modest winter peaked seasonality between 600 to 800 ng/m<sup>3</sup>. The fuel oil tracer vanadium on the other hand, is factor of two

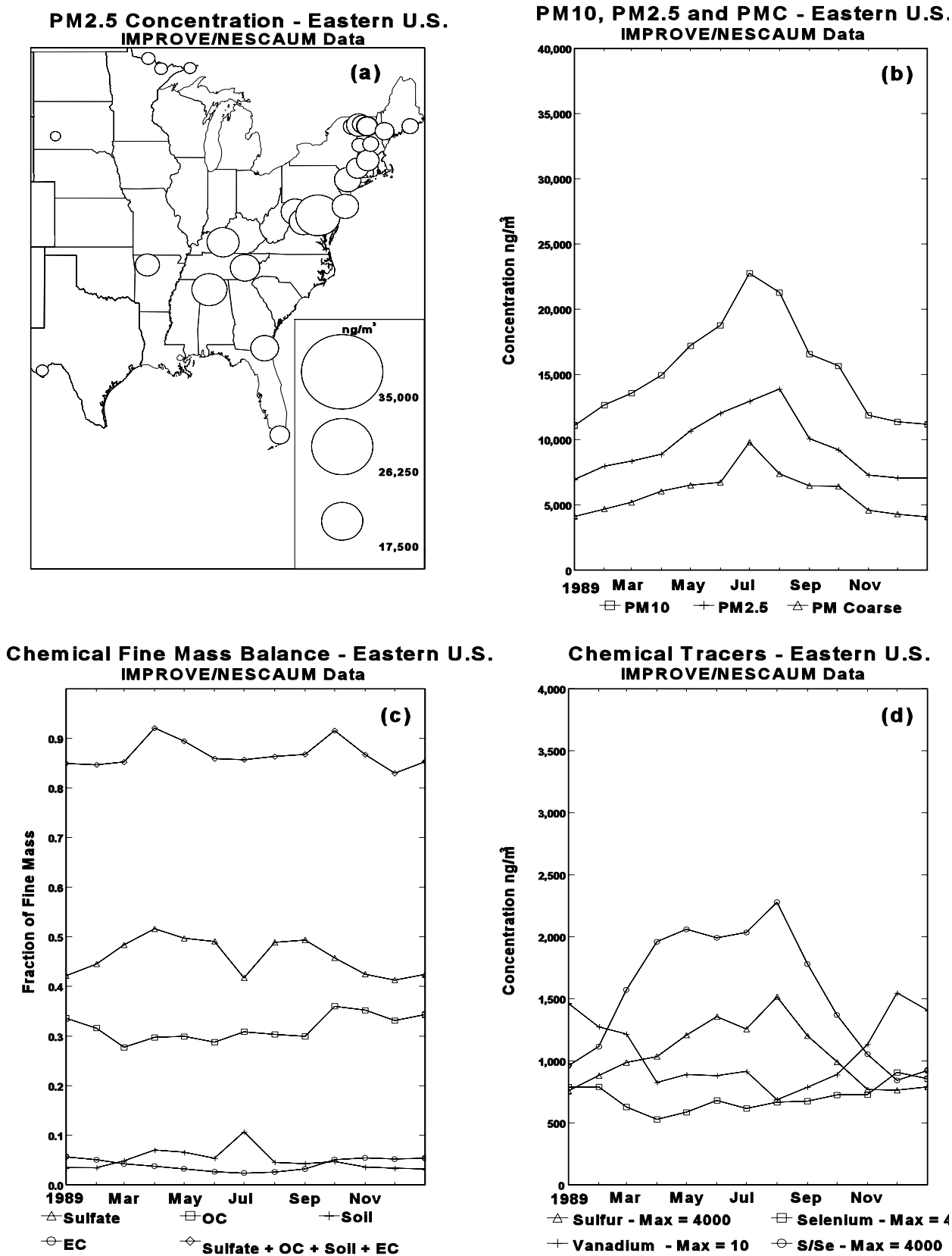


Figure 6-15. Seasonal pattern of nonurban aerosol concentrations for the eastern United States: (a) monitoring locations; (b) PM<sub>10</sub>, PM<sub>2.5</sub>, and PM Coarse (PMC); (c) sulfate, soil, organic carbon (OC), and elemental carbon (EC) fractions; and (d) tracers.



higher in the winter ( $1,500 \text{ ng/m}^3$ ) compared to the summer ( $750 \text{ ng/m}^3$ ). Evidently, the primary contribution from fuel oil is winter peaked. The S/Se ratio is about 1,000 in the winter, and it is over 2,000 in the summer months. This suggests the seasonality of secondary sulfate formation during the summer months.

### **Western United States**

The aggregated western U.S. aerosol seasonality is presented in Figure 6-16. The non-urban aerosol concentrations for  $\text{PM}_{10}$ ,  $\text{PM}_{2.5}$ , and  $\text{PM}_{\text{Coarse}}$  are well below the concentrations over the eastern United States (Figure 6-16b). The western United States differs from the eastern United States, having lower fine mass concentrations, which range between 3 to  $5 \mu\text{g/m}^3$ . The coarse mass concentration ( $4$  to  $8 \mu\text{g/m}^3$ ) and seasonality is similar over the East and the West. It is worth emphasizing, however, that these measurements are at remote national parks and wilderness areas in both East and West. The examination of monitoring data in urban areas and confined airsheds (Sections 6.4 and 6.5) reveals a highly textured pattern in space and time.

The fine particle chemical mass balance (Figure 6-16c) for the aggregated western United States shows the substantial contribution of organic carbon, which account for 30 to 45% of the fine mass. The higher organic carbon fraction occurs in the November through January season. Sulfates range between 20 to 25% throughout the year. Soil dust plays a prominent role in the western fine mass balance, contributing 20% in April through May, but declining to 5% by January. Elemental carbon ranges between 5% in the winter and 2 % during the summer. About 25% of the fine mass over the western United States is not accounted for by sulfates, soil, organic carbon, and elemental carbon. It is known that nitrates are major contributors to the fine particle mass in the South Coast Basin, as well as other western regions (White and Macias, 1987a; Chow et al., 1992a, 1993a, 1995a).

The concentration of the trace substances (Figure 6-16d) selenium and vanadium shows both low concentrations and weak seasonality. The sulfur concentrations are also less than half of the eastern U.S. values. The S/Se ratio is about 500 in the winter months and 1,000 during the summer. The lower S/Se ratios compared to those in the eastern U.S. are the result

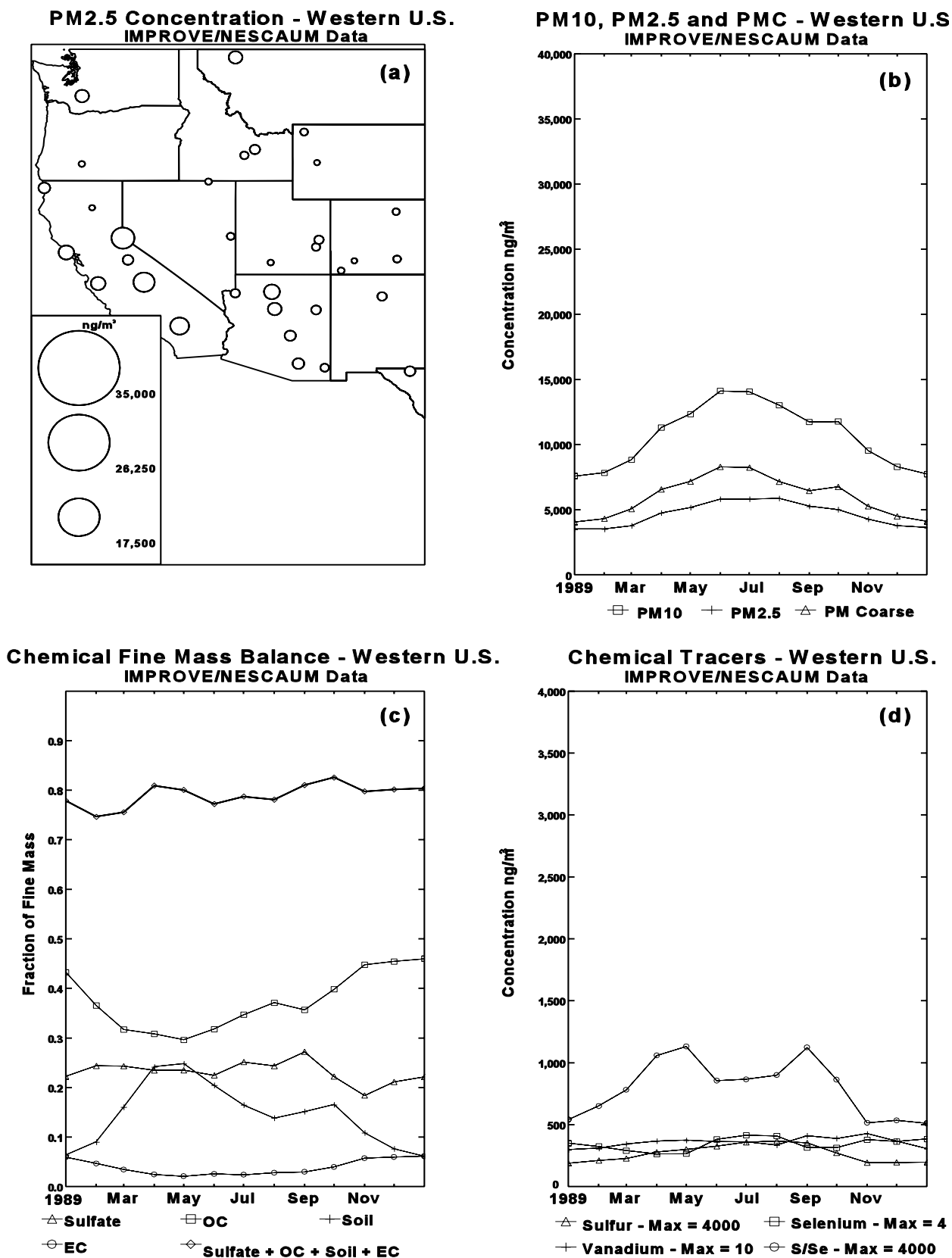


Figure 6-16. Seasonal pattern of nonurban aerosol concentrations for the western United States: (a) monitoring locations; (b) PM<sub>10</sub>, PM<sub>2.5</sub>, and PMCoarse (PMC); (c) sulfate, soil, organic carbon (OC), and elemental carbon (EC) fractions; and (d) tracers.

of selenium emitting coal-fired power plants not being the only sources of western U.S. sulfur. Smelters make a contribution to S but not Se in the atmosphere.

The above general discussion of national pattern of chemical and size dependence do not provide the more detailed spatial and temporal texture of the U.S. aerosol pattern discussed in the following sections. However, it provides the national scale gross features and serves as a broader context for the more detailed examinations.

#### **6.3.1.7 Background Concentrations of Particle Mass and Chemical Composition**

The concentration and chemical composition of background particulate matter can vary with geographic location, from monitoring site to monitoring site; with season of the year; and with meteorological conditions which affect the emissions and secondary production of biogenic or geogenic species to the background.

A number of types of background can be considered. These backgrounds include the following: (1) a "natural" background excludes all anthropogenic contributions. This background includes any natural sources contributing to the background for chemical species in North America or globally; (2) a background which excludes all anthropogenic sources within North America, but not from anthropogenic sources contributing to background from outside of North America; (3) a background which excludes the anthropogenic sources inside the United States, but not from elsewhere in North America; (4) a background which excludes anthropogenic sources from other regions into a specified region in the United States; (5) a background which would exclude all sources of particulate matter except those associated with a particular urban area. The two backgrounds directly relevant to the Criteria Document are backgrounds (1) and (2). The problems and limitations in obtaining reasonably accurate annual average and seasonal values for these backgrounds are discussed below. Backgrounds (4) and (5) can be more readily be obtained by measurements. These backgrounds are relevant to subsequent stages in the implementation process. The averaging period over which background levels are defined should also be stated. Annual and seasonal averages may be more appropriate for risk assessments but daily peak values may be more relevant for control strategy implementation.

More specifically, the term non-manmade is meant to encompass sources such as geogenic dust plumes and sea salt as well as biogenic sources. Biogenic sources include (a) combustion

products of biomass burning caused by lightning; (b) emissions of volatile sulfur compounds from marshes, swamps or oceans; (c) organic particulate matter formed by the atmospheric reactions of biogenic volatile organic compounds such as terpenes; and (d) particulate nitrates formed by the atmospheric reactions of  $\text{NO}_x$  emitted from soils. There is an intermediate class of sources associated with agricultural activities. These include biomass burning caused by human intervention and the addition of fertilizers to soils resulting in emissions of  $\text{NH}_3$  and  $\text{NO}_x$  (Section 5.2, 5.3).

Anthropogenic sources include vehicular and stationary sources which emit particles directly or precursors such as sulfur dioxide, nitrogen oxides, or those volatile organic compounds capable of reacting in the atmosphere to form organic particles. Stationary sources of primary particulate matter as well as sulfur oxides and nitrogen oxide precursors include fossil fuel power plants, while smelters are sources of primary particles and sulfur oxides. Vehicles emit primary particulate matter as well as nitrogen oxides and volatile organic compounds. Solvent usage, agricultural coatings, and many other industrial operations also may emit precursors or particulate matter. Wood burning for heating of homes is a source of organic carbon and elemental carbon (Section 5.2, 5.3).

The formation of sulfates from sulfur dioxide emitted by power plant plumes can occur over distances exceeding 300 km and 12 h of transport (Section 3.4.2.1). Nitric acid also can be formed in these plumes and it can be converted to ammonium nitrate, if sufficient ammonia is available to first neutralize the sulfate in plumes. Similar transport can occur in urban plumes. The transport distances in plumes depend on both formation rates of particles and their removal by deposition processes. However, the residence times of fine particles can be long. For example, if the dominant removal process is dry deposition, fine particles transported through a 1000 m deep mixed layer near the surface with deposition velocities of 1 to 0.1 cm/s have atmospheric residence times ranging from 1 to 11 days (Section 3.5.1, 3.5.3). When particles are trapped in a layer well aloft they may survive even longer periods. Therefore, transport distances of several hundred to several thousand kilometers are possible.

Direct evidence of such transport aloft is available from satellite monitoring of back scattered solar radiation. The most prominent plume is that of Sahara dust from West Africa (Section 6.2). This plume has been observed to extend during the spring and summer months to the east coast of the United States, especially over Florida (Figure 6-6). Ground level

measurements in Bermuda indicate that southeasterly winds bring high concentrations of soil- and crustal-related aerosols which appear to be from the Sahara desert (Wolff et al., 1986). Other large plumes exist, such as the Asian plume. However, the satellite observations do not indicate that it reaches the west coast of the United States (Figure 6-6).

Field measurements and modeling studies can be used as aids in the derivation of background values for aerosol constituents. Either approach is subject to considerable uncertainty and each has its own advantages and limitations. Field data would be the most logical choice if it could be shown to be completely free of anthropogenic influences originating within North America, i.e., background (2), (following the guidelines set out above for defining background levels unaffected by pollution sources within North America). A number of difficulties arise in interpreting field data for this purpose, namely: (a) there are very few tracers (e.g.,  $^{14}\text{C}$ ) which can be used to distinguish between anthropogenic and biogenic source categories of aerosol constituents; (b) multilayer trajectories should be used to identify source regions since layer-average trajectories may underestimate the geographic area contributing pollutants to the air mass sampled; (c) sampling must also be carried out for long enough periods to obtain statistically representative values over seasonal time scales. Determining the history of air parcels is difficult in locations subject to small scale circulations such as cumulus convection and land-sea or mountain-valley breezes. In addition, all small localized anthropogenic sources of particulate matter must be identified during sampling. Ideally, measurements should be carried out long enough for the measurements to be shown to be generally representative of the time period of interest e.g., seasonal average, annual average.

Alternatively, models which include only natural sources and anthropogenic sources located outside North America could be used. Their utility is limited by inadequacies in model formulation, such as grid spacing and knowledge of the strengths, locations, and variability of various sources. Since a large fraction of particulate matter is secondary, uncertainties in the chemistry of precursor gases will play a large role in determining the uncertainty of the final results. These uncertainties are especially large for the yield of aerosol produced by the oxidation of biogenic hydrocarbons as pointed out in Chapters 3 and 5. Uncertainties in the chemistry of  $\text{NO}_x$  and  $\text{SO}_2$  are also important in that they affect estimates of the yield of aerosol products versus the deposition of intermediate species.

Trijonis (1982, 1991) has attempted to estimate  $PM_{2.5}$  and  $PM_{10}$  concentrations corresponding to background (1), the "natural" background. His approach was to obtain concentration values only from those biogenic and geogenic sources which are at or below those possibly associated with preindustrial conditions over North America. Annual average concentrations of the chemical species in particulate matter were estimated for the eastern United States and for mountain/desert regions of the western United States. Seasonal "natural" background concentration values were not estimated. The annual average concentrations of fine particles were estimated separately for sulfates; as  $NH_4HSO_4$ , nitrates; as  $NH_4NO_3$ ; organic carbon; elemental carbon; soil dust and water (Trijonis, 1982, 1991). In the later work, coarse particle concentration values were also estimated (Trijonis, 1991). In addition, in the later work, it was emphasized that the concentration values proposed can have error factors ranging from 1.5 to 3 for individual chemical species in particulate matter.

In the earlier work (Trijonis, 1982), a fine particle "natural" background for the eastern United States is estimated at  $5.5 \pm 2.5 \mu g/m^3$ . Excluding water, the background value would be  $4 \pm 2 \mu g/m^3$  with the largest contribution,  $2 \mu g/m^3$ , from organic carbon. In the later estimates (Trijonis, 1991), a fine particle "natural" background for the eastern United States of  $3.3 \mu g/m^3$  is estimated. Excluding water, this background would be  $2.3 \mu g/m^3$  with  $1.5 \mu g/m^3$  associated with organic carbon. A separate estimate is given for the fine particle "natural" background over the mountain/desert regions of the western United States of  $1.2 \mu g/m^3$ . Excluding water, this background would be  $1 \mu g/m^3$  with  $0.5 \mu g/m^3$  associated with organic carbon. The coarse particle "natural" background for both the eastern and western United States is estimated at  $3 \mu g/m^3$ .

Fernam et al. (1981) also estimated "natural" background concentrations for  $PM_{2.5}$  constituents in the eastern United States during summer. They estimated natural contributions to sulfate of  $0.5\text{-}1.9 \mu g/m^3$ , to organic carbon of  $3.7 \mu g/m^3$ , and to crustal material of  $1.7 \mu g/m^3$ .

To obtain these "natural" background estimates, a wide range of approaches are used varying from natural  $SO_2$  and  $NO_x$  emissions inventories to  $SO_4^{2-}$ ,  $NO_3^-$  and elemental carbon concentration measurements in remote locations in the northern and southern hemispheres. Carbon isotope ratios and organic composition measurements for organic components are used from several sites in the southwestern United States.

Results of three-dimensional models that could be used to estimate each of the five background levels for all the major categories of aerosol composition listed above are not available. Lioussé et al. (1996) have performed three-dimensional chemical tracer model simulations of the global distribution of elemental and organic carbon. Background values assuming only natural sources (background 1) were also calculated. Average organic carbon concentrations calculated for the month of July were all less than  $1 \mu\text{g}/\text{m}^3$  in the United States. These calculations were made assuming a 5% yield of secondary organic carbon from the oxidation of terpenes (cf. Section 5-3).

Another approach is to use results from rural/remote sites in national parks, wilderness areas and national monuments from the IMPROVE monitoring measurements. Results for the period between March 1988 and February 1991 have been published (Malm et al., 1994). The tabulations of results are given on an annual average basis for individual IMPROVE sites and on a seasonal basis by IMPROVE subregion for fine mass; sulfate, as  $(\text{NH}_4)_2\text{SO}_4$ ; nitrate, as  $\text{NH}_4\text{NO}_3$ ; organic and elemental carbon; fine soil and coarse mass. These measurements do not differentiate between anthropogenic and non-anthropogenic contributions and do not stratify measurements values by wind direction or by use of trajectories representing various air masses (Malm et al., 1994). However, a large set of measurements, including seasonal measurements, are provided at a substantial number of rural/remote sites, especially in the western United States.

In stratifying the IMPROVE results a problem arises because the Colorado plateau "subregion" with seven sites straddles the boundary between the southwest and northwest used subsequently (Figure 6-28). Four of the sites are north of the boundary in Utah and Colorado and three of the sites are south of the boundary in Arizona and New Mexico. The authors place the Colorado plateau in the southwest for purposes of a fine mass composition budget (Malm et al., 1994). Since they assign only one other subregion, Sonora desert, with two sites to the southwest, the method of assigning sites can significantly affect the resulting estimates of regional fine mass concentrations. This problem can be avoided for the annual average values which are shown by individual sites, but not for the seasonal values which are lumped by subregion. This lumping also requires deciding whether a subregion with five sites, central Rocky Mountains, should be given the same or five times the weight of the other subregions in the northwest with only one or two sites each. For the annual average values given in Table 6-2

the sites are assigned consistent with the division between the northwestern and southwestern regions shown in Figure 6-28, excluding three sites in the northern California coastal mountains considered separately. A transitional region between the western mountains and deserts and the eastern United States has been considered consisting of five sites in three subregions from West Texas (2), to South Dakota (1) up to the Boundary Waters subregion (2) near the Canadian border. In addition, the result for particulate matter from the Appalachian subregion (2) are given. Previous measurements of particulate matter at sites in the eastern mountains are available (Stevens et al., 1980); Pierson et al., 1980b; Wolff et al., 1983). The measurements listed in Table 6-2 include  $PM_{2.5}$  sulfate, as  $(NH_4)_2SO_4$ , organic carbon, and  $PM_{(10-2.5)}$ .

The annual average  $PM_{2.5}$  increases substantially from west to east in Table 6-2 from a value of  $3.55 \mu g/m^3$  in the northwestern United States to  $10.91 \mu g/m^3$  in the Appalachian mountains. The annual average  $(NH_4)_2SO_4$  concentration increases even more substantially from west to east from a value of  $0.88 \mu g/m^3$  in the northwestern United States to  $6.33 \mu g/m^3$  in the Appalachian Mountains. The lowest annual average organic carbon concentration of  $1.38 \mu g/m^3$  occurs in the southwestern United States and increases to  $2.97 \mu g/m^3$  in the Appalachian Mountains. A smaller range of concentrations occurs for organic carbon from west to east than for  $PM_{2.5}$  and  $(NH_4)_2SO_4$ . The  $(NH_4)_2SO_4$ , as a percentage of  $PM_{2.5}$ , increases into the transitional region and the Appalachian Mountains from as low as 25% of the  $PM_{2.5}$  at sites in the northwestern United States up to 58% at sites in the Appalachian Mountains. Conversely, organic carbon decrease as a percentage of  $PM_{2.5}$  from 46% at sites in northwestern United States down to 27% in the Appalachian Mountains. Within the



**TABLE 6-2. ANNUAL AVERAGE CONCENTRATIONS AND  
CHEMICAL COMPOSITION FROM IMPROVE MONITORING SITES**

	No. of	Annual Average Concentrations, $\mu\text{g}/\text{m}^3$ and Composition				
			(NH <sub>4</sub> ) <sub>2</sub> SO <sub>4</sub> /%	Organics/	PM	
Northwest <sup>a</sup>	15	3.55	0.88/25	1.63/46	4.46	8.0
Southwest <sup>b</sup>	5	3.91	1.28/33	1.38/35	5.62	9.5
California Coastal Mountains <sup>c</sup>	3	4.99	1.41/28	1.95/39	8.85	13.8
Transitional Region <sup>d</sup>	5	5.15	1.97/38	2.01/39	6.54	11.7
Appalachian Mountains <sup>c</sup>	2	10.91	6.33/58	2.97/27	6.24	17.2

<sup>a</sup>Cascades (1), central Rocky Mt. (5), Great Basin (1), N. Rocky Mt. (1), Sierra Nevada (1), Sierra Humboldt (2), and Colorado Plateau (4)

<sup>b</sup>Colorado Plateau (3), Sonora Desert (2)

<sup>c</sup>Same as subregion

<sup>d</sup>Western Texas (2), northern Great Plains (1), Boundary Waters (2).

western United States there are somewhat higher percentages of (NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub> and lower percentages of organic particles in the southwestern United States than in the northwestern United States. (NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub> plus organic carbon account for from 67% to 85% of PM<sub>2.5</sub>, with the higher percentages at IMPROVE sites east of the Rocky Mountains (Table 6-2).

Compared to the estimates discussed by Trijonis (1982, 1991) for "natural" background, PM<sub>2.5</sub> values in the western United States of 1  $\mu\text{g}/\text{m}^3$ , the average measured concentrations of PM<sub>2.5</sub> in the northwestern and southwestern United States of 3.55  $\mu\text{g}/\text{m}^3$  and 3.91  $\mu\text{g}/\text{m}^3$  suggest anthropogenic contributions. The IMPROVE measurements are likely to include anthropogenic contributions from sources within North America (background 3). Even the lowest annual average PM<sub>2.5</sub> value in the contiguous United States of 2.5  $\mu\text{g}/\text{m}^3$  at Bridger Wilderness Area, WY, is over twice the "natural" background. The Denali NP in Alaska has an average annual PM<sub>2.5</sub> of 2  $\mu\text{g}/\text{m}^3$  (Malm et al., 1994). The organic carbon concentrations measured there are somewhat closer to the estimated "natural" background in the western mountains/desert of 0.5  $\mu\text{g}/\text{m}^3$  (Trijonis, 1991). However, average annual concentrations in the northwestern and southwestern United States are higher with values of 1.63  $\mu\text{g}/\text{m}^3$  and 1.38  $\mu\text{g}/\text{m}^3$ . The annual average values at several IMPROVE monitoring sites in the Rocky Mountains are near 1  $\mu\text{g}/\text{m}^3$ , while the Denali NP in Alaska has an average annual organic carbon concentration of

0.85  $\mu\text{g}/\text{m}^3$ . These latter organic carbon concentration values are at the two fold upper limit of uncertainty in the estimate of "natural" background. On the other hand, the  $(\text{NH}_4)_2\text{SO}_4$  concentrations measured in the west are far above the "natural" background for  $(\text{NH}_4)_2\text{SO}_4$  of 0.1  $\mu\text{g}/\text{m}^3$  (Trijonis, 1991). The lowest measured annual average  $(\text{NH}_4)_2\text{SO}_4$  at several sites are near 0.5  $\mu\text{g}/\text{m}^3$ . For  $\text{PM}_{(10-2.5)}$ , the annual average concentrations in the northwestern and southwestern United States of 4.46  $\mu\text{g}/\text{m}^3$  and 5.62  $\mu\text{g}/\text{m}^3$  are within the two fold upper limit of uncertainty in the estimate of "natural" background. At a number of individual sites, annual average  $\text{PM}_{(10-2.5)}$  concentrations are 3  $\mu\text{g}/\text{m}^3$  to 3.5  $\mu\text{g}/\text{m}^3$ , close to the estimated "natural" background. Therefore, the largest deviations from the "natural" background estimates for a major component occur for  $(\text{NH}_4)_2\text{SO}_4$ .

Comparisons of the measured concentration values in the "transitional" area of the eastern United States, using sites from west Texas to the Boundary Waters, find that the average annual concentrations for  $\text{PM}_{2.5}$  of 5.15  $\mu\text{g}/\text{m}^3$ ;  $(\text{NH}_4)_2\text{SO}_4$  of 1.97  $\mu\text{g}/\text{m}^3$ ; organic carbon of 2.01  $\mu\text{g}/\text{m}^3$  and  $\text{PM}_{(10-2.5)}$  of 6.54  $\mu\text{g}/\text{m}^3$  (Table 6-2) usually are well above the estimates of "natural" background in the eastern United States (Trijonis, 1991) for  $\text{PM}_{2.5}$  of 2.3  $\mu\text{g}/\text{m}^3$ ;  $(\text{NH}_4)_2\text{SO}_4$  of 0.2  $\mu\text{g}/\text{m}^3$ ; organics of 1.5  $\mu\text{g}/\text{m}^3$ ; and  $\text{PM}_{(10-2.5)}$  of 3  $\mu\text{g}/\text{m}^3$ . As in the western United States, the measured  $(\text{NH}_4)_2\text{SO}_4$  concentration values are far above the "natural" background value, while the measured concentrations of organics are well within the two fold uncertainty in the "natural" background value.

Another source of lower  $\text{PM}_{10}$  concentrations are rural/remote AIRS monitoring sites. Based on 1993 measurements, the lowest values of  $\text{PM}_{10}$  are as follows: Rosebud Co., MT (maximum of 10  $\mu\text{g}/\text{m}^3$ , annual mean of 4.5  $\mu\text{g}/\text{m}^3$ ); Campbell Co., WY (maximum of 15  $\mu\text{g}/\text{m}^3$ , annual mean of 7.0  $\mu\text{g}/\text{m}^3$ ); and Washington Co., ME (maximum of 23  $\mu\text{g}/\text{m}^3$ , annual mean of 8.8  $\mu\text{g}/\text{m}^3$ ). These  $\text{PM}_{10}$  values agree within a factor of two with the estimated "natural" background  $\text{PM}_{10}$  in the western United States of 4  $\mu\text{g}/\text{m}^3$ , and in the eastern United States of 5.3  $\mu\text{g}/\text{m}^3$  (Trijonis, 1991).

Seasonal variations in particulate matter are also important and have been considered. The source used for these seasonal values in particulate matter is the IMPROVE monitoring network (Malm et al., 1994). Because the seasonal values are reported only by IMPROVE subregions, there is no good approach to averaging values from differing numbers of sites within the varying geographical extent of IMPROVE subregions. Therefore, the values of annual average, summer

and winter values for  $PM_{2.5}$ ,  $(NH_4)_2SO_4$ , organic carbon, and  $PM_{(10-2.5)}$  are listed for a number of IMPROVE subregions (Table 6-3).

**TABLE 6-3. ANNUAL SUMMER AND WINTER CONCENTRATIONS FROM IMPROVE MONITORING SITES<sup>a</sup>**

Subregion	Region of U.S.	No of Sites	Seasons of the Year	$PM_{2.5}$	$(NH_4)_2SO_4$	Organics	PM Coarse
Central Rockies	NW	5	annual	3.3	0.8	1.5	4.8
			summer	4.8	1.0	2.4	7.5
			winter	2.0	0.5	0.9	3.0
Colorado Plateau	NW-SW	7	annual	3.4	1.1	1.2	4.7
			summer	4.1	1.3	1.6	6.4
			winter	2.9	0.9	1.1	3.2
Coastal Mountains	NW	3	annual	5.0	1.4	1.9	8.9
			summer	4.5	1.9	1.4	10.7
			winter	5.6	0.9	2.3	7.7
Sonora Desert	SW	2	annual	4.4	1.5	1.5	6.0
			summer	5.6	2.1	1.8	7.6
			winter	3.2	1.2	1.1	3.3
West Texas	Transitional to east	2	annual	5.4	2.1	1.5	7.5
			summer	6.6	2.5	1.7	7.4
			winter	3.6	1.5	1.1	5.1
Northern Great Plains	Transitional to east	1	annual	4.5	1.5	1.5	6.3
			summer	5.6	1.8	2.2	9.7
			winter	3.4	1.2	1.1	3.9
Boundary Waters	Transitional to east	2	annual	5.3	2.0	2.1	5.7
			summer	6.2	2.2	3.1	8.2
			winter	5.2	2.0	1.4	3.2
Appalachian Mountains	Eastern U.S.	2	annual	10.9	6.3	3.0	6.2
			summer	16.6	10.5	4.4	11.2
			winter	6.5	3.0	2.0	3.1

<sup>a</sup> From Malm et al., 1994.

Annual average concentration almost always are intermediate between the summer and winter concentration of particulate matter listed in Table 6-3. With a few exceptions, the

summer concentrations are higher than winter concentrations. The exceptions are the higher winter concentrations for  $PM_{2.5}$  and organics in the coastal mountains. Ratios of summer to winter concentrations can equal or exceed two for all listed particulate components in both the central rockies subregion and the Appalachian Mountains. The summer to winter concentration ratios for  $PM_{2.5}$  are within the 1.5 to 2.5 range except for the coastal mountains and Boundary Waters subregion. The summer to winter concentration ratios for PM coarse equal or exceeds two except for the coastal mountains. Therefore, in most rural remote sites in IMPROVE subregions summer concentrations of particulate matter substantially exceed winter concentrations. However, it must be emphasized that it is not appropriate to extrapolate these results obtained at IMPROVE sites in 1988 to 1991 to other sites or even to other years of monitoring at IMPROVE sites.

Within the continental United States, there are measurements of particulate mass and chemical composition under conditions identified as "clean" background conditions (Wolff et al., 1983). These are based on 7 days of measurements during the summer of 1978 at a site 40 km northwest of Pierre, South Dakota and 18 days during the summer of 1979 at a site 15 km north of the Gulf Coast, near Abbeville, LA. At the South Dakota site the small variations in anthropogenic pollutants observed was attributed to a lack of any major pollution sources along the trajectories. In contrast, at the Louisiana site the days were stratified into "clean" days when the air had passed over the Gulf of Mexico for several days and much more polluted episode days when the maritime air was modified by air which had undergone transport from the midwestern and northeastern United States.

Fine particle mass on "clean" days averaged 11 to 13  $\mu\text{g}/\text{m}^3$  and coarse mass between 9 and 19  $\mu\text{g}/\text{m}^3$  at the two sites. The total mass averaged between 21 and 32  $\mu\text{g}/\text{m}^3$ . Organic carbon at both sites was the most important fine particle species averaging 4 to 8  $\mu\text{g}/\text{m}^3$  (organic mass multiplied by 1.2 to include H and O), while sulfate averaged 3  $\mu\text{g}/\text{m}^3$ .

At the closest IMPROVE site, the Badlands National Monument, SD in the northern great plains subregion (Table 6-3), for the summers of 1988 and 1989 (Malm et al., 1994) the concentrations were  $PM_{2.5}$ , 5.6  $\mu\text{g}/\text{m}^3$ ;  $(\text{NH}_4)_2\text{SO}_4$ , 1.8  $\mu\text{g}/\text{m}^3$ ; organic carbon, 2.2  $\mu\text{g}/\text{m}^3$  and  $PM_{(10-2.5)}$ , 9.7  $\mu\text{g}/\text{m}^3$ . These concentration values are substantially lower than those obtained at the site 40 km northwest of Pierre, SD in the summer of 1978 as follows:  $PM_{2.5}$ , 13  $\mu\text{g}/\text{m}^3$ ,  $(\text{NH}_4)_2\text{SO}_4$ , 3.2  $\mu\text{g}/\text{m}^3$ ; organic carbon 3.8  $\mu\text{g}/\text{m}^3$  and  $PM_{(10-2.5)}$ , 19  $\mu\text{g}/\text{m}^3$ .

There are several reasons for the differences between the "clean" values and the IMPROVE values. (a) The measured background varies from year to year and site to site. (b) Precipitation periods were excluded by Kelly et al. (1982) and Wolff et al. (1983), but the IMPROVE monitoring network measurements include all weather conditions. All other conditions being the same, the inclusion of precipitation events in the IMPROVE measurements probably biases the results low because of rain-out of particulate matter. (c) On the other hand, the presence of material from anthropogenic sources probably biases the results high. Wolff et al. (1983) used trajectory analyses to exclude periods with intrusions of polluted air from their analysis. This was not done with the IMPROVE results. However, the layer-averaged trajectories used by Wolff et al. (1983) may have underestimated the mixing of air parcels from surrounding geographical areas leading to an underestimate of the potential for anthropogenic contributions. The exact causes for the differences between these two types of "background" estimates cannot be quantitated from available data.

For sulfate, it is possible to make a limited comparison with measurements at rural sites outside of St. Louis with air flow from the northwest during the third quarters of 1975 and 1976 (Altshuller, 1987), background 5. The average third quarter sulfate concentrations at these sites for these two years was  $7 \mu\text{g}/\text{m}^3$ , a substantially higher sulfate concentration than in South Dakota (Wolff et al., 1983), but lower than measured in other wind directions. These measurements outside of St. Louis also indicate substantially lower sulfate concentrations during the first and fourth quarters of 1975, 1976, and 1977 averaging  $3.4 \mu\text{g}/\text{m}^3$ , comparable to the third quarter sulfate concentrations in South Dakota.

It is important to emphasize that the "background" for particulate matter moving toward cities along the east coast over the Great Smoky Mountains (Stevens et al., 1980); the Allegheny Mountains (Pierson et al., 1980b) and the Blue Ridge Mountains (Wolff et al., 1983), background 4, are much higher than for the "clean" air days in South Dakota and Louisiana. For example, the fine particle matter at the Blue Ridge Mountain site in July and August 1980 with trajectories from the midwest source areas and the Tennessee Valley source area averaged 27 and  $24 \mu\text{g}/\text{m}^3$ , approximately twice the values under "clean" air conditions in South Dakota and Louisiana (Wolff et al., 1983). The sulfate concentrations for these two trajectory directions averaged 14 and  $9 \mu\text{g}/\text{m}^3$ , with sulfate substantially exceeding organic carbon. This result is a reversal in the chemical composition under the "clean" air conditions in South Dakota and

Louisiana, but more consistent with the chemical composition under "episodic" conditions in Louisiana when the sulfate concentration averaged  $20 \mu\text{g}/\text{m}^3$  and the organic carbon averaged  $15 \mu\text{g}/\text{m}^3$ .

Because of the repeated occurrence of  $(\text{NH}_4)_2\text{SO}_4$  concentrations far above "natural" background even at rural/remote sites, this aspect justifies additional consideration.

A low contribution of natural sources of gaseous sulfur (both terrestrial and marine) occurs in the eastern United States (Trijonis, 1991). However, a more detailed consideration of the contribution of natural sources of gaseous sulfur indicates wide variations over the United States (Placet, 1991). The following estimates for the ratios of total natural gaseous sulfur to total anthropogenic gaseous sulfur by region (Placet, 1991) are as follows: northeast, 0.01; southeast, 0.03; west gulf, 0.03; southwest, 0.12; northwest, 0.19. The corresponding ratios for coastal areas are higher with an estimate of 0.52 for the California coastal areas. If these ratios are converted to ratios of total natural gaseous sulfur to total gaseous sulfur, the ratios would be 0.11 in the southwest and 0.16 in the northwest. If the following assumptions are made (a) both natural and anthropogenic sulfur are converted to  $(\text{NH}_4)_2\text{SO}_4$  to about the same extent; (b) the concentrations of natural  $(\text{NH}_4)_2\text{SO}_4$  can be obtained by multiplying the above ratios by the measured  $(\text{NH}_4)_2\text{SO}_4$  concentrations, the natural sulfur concentrations in the southeast would range from 0.1 to  $0.15 \mu\text{g}/\text{m}^3$  and in the northwest from  $0.08 \mu\text{g}/\text{m}^3$  to  $0.2 \mu\text{g}/\text{m}^3$ .

A more detailed consideration of the contribution of natural gaseous sulfur at sites near the Pacific coast is available (Kreidenweis, 1993). In particular, comparisons with measured  $(\text{NH}_4)_2\text{SO}_4$  concentrations were made at the Crater Lake National Park in southwestern Oregon with estimates of natural  $(\text{NH}_4)_2\text{SO}_4$  concentrations. The measured annual average concentration at this site of  $(\text{NH}_4)_2\text{SO}_4$  was  $0.5 \mu\text{g}/\text{m}^3$  and an average "low" concentration was approximately  $0.13 \mu\text{g}/\text{m}^3$  (Kreidenweis, 1993). This latter value can be compared with several estimates of natural  $(\text{NH}_4)_2\text{SO}_4$  concentration based on the following approaches (a) a natural source column burden between  $35$  to  $50^\circ$  north of  $0.05$  to  $0.15 \mu\text{g}/\text{m}^3$ ; (b) a Pacific natural source column estimate between  $35$  to  $50^\circ$  N of  $0.18 \mu\text{g}/\text{m}^3$  and (c) a 3 D model value of  $0.14$  to  $0.28 \mu\text{g}/\text{m}^3$ . Other approaches gave higher possible values for natural  $(\text{NH}_4)_2\text{SO}_4$  (a) "clean" rainfall sulfate concentrations of  $0.1$  to  $0.5 \mu\text{g}/\text{m}^3$  and (b) another 3-D model value of  $0.6 \mu\text{g}/\text{m}^3$ . These comparisons results in a wide range of annual average values of  $(\text{NH}_4)_2\text{SO}_4$  from less than  $0.1 \mu\text{g}/\text{m}^3$  to less than  $0.5 \mu\text{g}/\text{m}^3$  (Kreidenweis, 1993).

Even an upper limit value for natural  $(\text{NH}_4)_2\text{SO}_4$  of  $0.5 \mu\text{g}/\text{m}^3$  would be a third to a half of the measured  $(\text{NH}_4)_2\text{SO}_4$  at IMPROVE sites near the Pacific Coast (Malm et al., 1994). Further inland, at interior western sites, the marine sources of natural sulfur should make an even smaller contribution to the measured concentrations of  $(\text{NH}_4)_2\text{SO}_4$ . Comparison of these  $(\text{NH}_4)_2\text{SO}_4$  with the estimates based on regional sulfur inventories (Placet, 1991) of 0.08 to  $0.2 \mu\text{g}/\text{m}^3$  would indicate a significant anthropogenic contribution even at relatively remote western IMPROVE sites. This result suggests that background 3 may have a substantial contribution from anthropogenic sulfur sources in North America.

As a summary to the discussion in Section 6.3.1.7, the estimated lower limit and upper limit background concentrations for  $\text{PM}_{10}$  and  $\text{PM}_{2.5}$  are given on an annual average basis and for winter and summer for the western and eastern United States in Table 6-4.

**TABLE 6-4. SUMMARY OF ANNUAL AND SEASONAL AVERAGE RANGES OF BACKGROUND CONCENTRATION LEVELS OF  $\text{PM}_{10}$  AND  $\text{PM}_{2.5}$**

PM	Annual or Seasonal	Concentrations, $\mu\text{g}/\text{m}^3$	
		Western United States	Eastern United States
$\text{PM}_{10}$	Annual average	4 - 8	5 - 11
$\text{PM}_{2.5}$	Annual average	1 - 4	2 - 5
$\text{PM}_{10}$	Winter	4 - 6	5 - 8
$\text{PM}_{2.5}$	Winter	1 - 3	2 - 4
$\text{PM}_{10}$	Summer	4 - 12	5 - 14
$\text{PM}_{2.5}$	Summer	1 - 5	2 - 6

The lower limit concentrations are based on the "natural" background midrange concentrations discussed (Trijonis 1991). There are error factors associated with the chemical species used to obtain these concentrations range from 1.5 to 3.

The upper limit concentrations are based on measured concentrations from IMPROVE monitoring sites (Malm et al., 1994). The  $\text{PM}_{2.5}$  concentrations are the sum of concentrations measured for individual chemical species. As noted earlier in Section 6.3.1.7, these measured concentrations can include some anthropogenic source contributions within North America.

Therefore, the upper limit concentrations represent conservative values for the upper end of the background concentration type.

To obtain the upper limit concentrations, the averages were obtained from the concentrations for nine subregions in the western United States giving each region equal weight and also weighing the contribution of each subregion by the number of sites in the subregion. The median values were also obtained. For the eastern United States, the averages were obtained from the concentrations for three subregions in the "transitional" region. For the annual average from 23 individual sites in the western United States and 5 sites in the "transitional" region (Table 6-3). The resulting values for upper limit concentrations were closely clustered usually with a  $1 \mu\text{g}/\text{m}^3$  range. Within these values, the lower whole value concentration was listed in Table 6-4.

As a supplement to the data collected in the IMPROVE/NESCAUM networks, seasonal and annual average  $\text{PM}_{10}$  concentrations were also taken from AIRS (1990 - 1995). Four inhabited areas with the lowest annual average  $\text{PM}_{10}$  concentrations were chosen in areas without nearby IMPROVE/NESCAUM sites. Annual, summer, and winter averages for Penobscot Co., ME (11.1, 13.8, and  $10.0 \mu\text{g}/\text{m}^3$ ); Marquette, MI (11.2, 15.5, and  $7.0 \mu\text{g}/\text{m}^3$ ); Mercer Co., ND (11.7, 12.9, and  $10.6 \mu\text{g}/\text{m}^3$ ); and Lakeport, CA (11.6, 14.3, and  $10.0 \mu\text{g}/\text{m}^3$ ) all fall within the upper limits set for  $\text{PM}_{10}$  shown in Table 6-4. All areas exhibit summertime maxima and wintertime minima. The similarity of these results to the upper limits shown in Table 6-4 suggests an anthropogenic component to those upper limits, since the AIRS values were obtained in inhabited areas.

Again, it should be mentioned that seasonal or annual average "background" values presented above will likely underpredict 24-hour maximum "background" values. Ambient data could be used to estimate 24-hour maximum values, but their use is subject to considerable uncertainty because of possible anthropogenic inputs.

### **6.3.2 Urban National Aerosol Pattern—Aerometric Information Retrieval System**

The urban monitoring network is operated by state and local agencies as mandated by the Clean Air Act. The data from this network are used to determine exceedences above the particulate matter standards. Federal regulations also require that these monitoring data be



submitted to the EPA Aerometric Information and Retrieval System (AIRS). In what follows, AIRS  $PM_{10}$  refers to the  $PM_{10}$  mass concentration extracted from the AIRS database. The AIRS database is a useful resource for analyzing trends and concentration patterns, and relationships between the fine, coarse, and  $PM_{10}$  components of the atmospheric aerosol (Husar and Frank, 1991; Husar and Poirot, 1992).

The national average AIRS concentrations were calculated utilizing all of the available data since the beginning of 1985, when less than 100 monitoring stations were operational (Figure 6-17). Since that time, the number of monitoring stations has risen to more than 1,300 (Figure 6-17). The implications of the changing stations density to the above described national  $PM_{10}$  trend is not well studied. The emergence of new stations appeared to be in rough proportion to the final station density shown in Figure 6-17. In other words, in 1985, the national coverage had a pattern similar to 1994, except less dense. Changes in sampling equipment and monitoring protocols are also possible causes of systematic errors in the reported spatial pattern and trends.

The AIRS  $PM_{10}$  database reports the concentrations every sixth day for a 24-h sampling period, synchronously over the entire country. The sample duration is one day which, over the long run, provides the concentration distribution function of daily samples. For determination of the effects (human health, visibility, acid deposition) the concentration has to be known at the specific location where the sensitive receptors reside. Also the concentrations have to be known at a short (e.g., daily) time scale, as well as over the long term.

In order to characterize the one day-scale temporal variation over a given region, the entire available data aggregated over the entire region for each monitoring day are plotted as

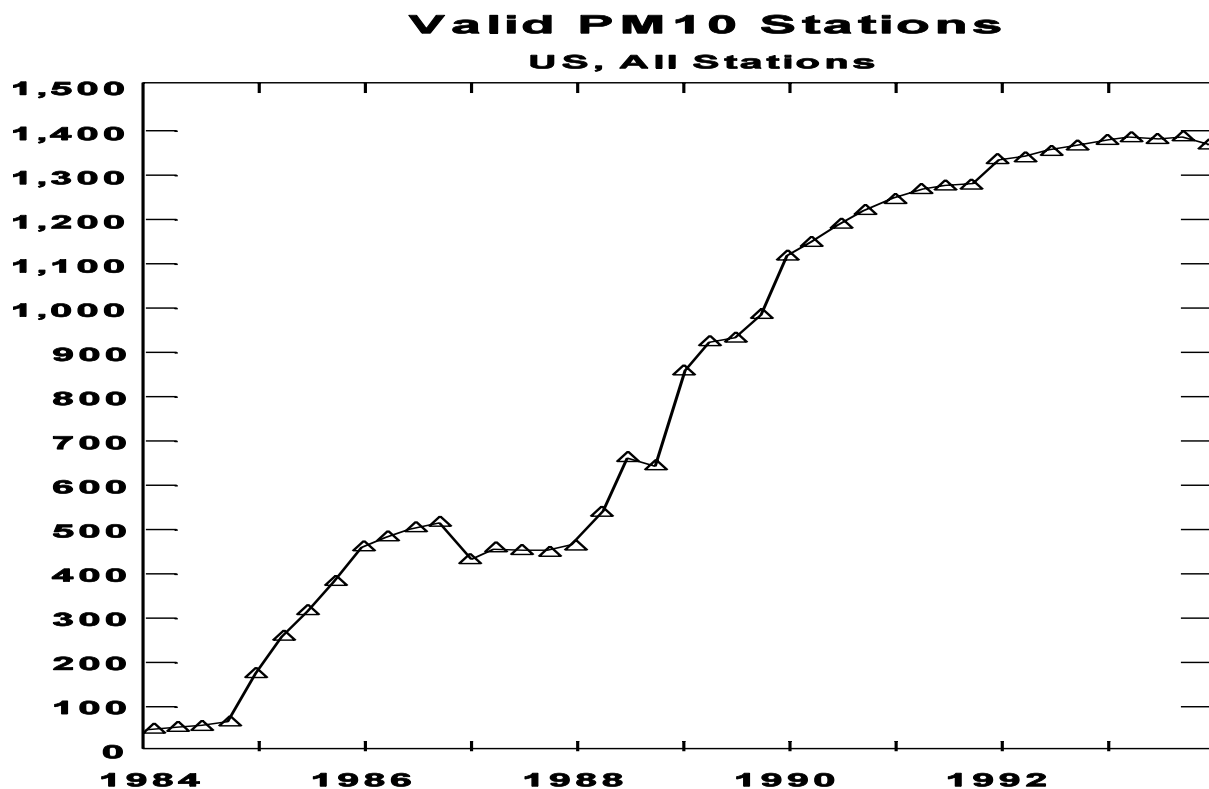


Figure 6-17. Trend of valid PM<sub>10</sub> monitoring stations in the AIRS database.

time series. It is recognized that during the other five non-monitored days, the concentrations may be different from the reported value. The six day sample increment ensures that both weekday and weekend data are properly taken into account.

The AIRS PM<sub>10</sub> stations are mostly in urban areas but some suburban and nonurban sites are also reported. The analysis presented in this section is based on PM<sub>10</sub> and PM<sub>2.5</sub> data retrieved from AIRS in October 1994.

The results of AIRS PM<sub>10</sub> aerosol pattern analysis are presented in quarterly contour maps, as well as seasonal time charts. For valid monthly and quarterly aggregation, it was required to have at least two samples a month, and six samples per quarter. For the seasonal maps all the available data between 1985 to 1994 were used.

The seasonal contour maps also show the location of the PM<sub>10</sub> monitoring sites. The size of the rectangle at each site is proportional to the quarterly average PM<sub>10</sub> concentration